Electronic Structure of $CeRu_2Si_2$

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INTRODUCTION

The large correlation energies of *f*-states in mixed-valent and heavy fermion rare-earth and actinide compounds results in a challenging interplay of atomic and electron gas physics, generally understood to involve hybridization between localized *f*-orbitals and other band-like *s-p-d* states of the solid. For a single material, some properties, e.g. Kondo effects, can be described by the impurity Anderson model, and yet the *f*-electrons contribute to the heavy-mass Fermi surface (FS) [1,2]. The different theoretical models make specific predictions about changes in the FS topology resulting from the introduction of highly-correlated *f*-states as compared to the non-*f* bandstructure, *e.g.* that in certain circumstances the overall FS topology appears to be largely determined by the underlying *s-p-d* band structure in the absence of hybridization to the *f*-states [3].

Hence it is important to study the detailed differences between similar systems with varying *f*-occupation, both experimentally and theoretically, in order to elucidate the effects of *f*-correlation. The XRu_2Si_2 system with X=(La, Ce, Th, U) is a favorable isostructural series for characterization with only the 4*f* or 5*f* occupation being varied. $CeRu_2Si_2$ is a $4f^1$ heavy fermion system (γ=350 mJ/mol-K^2) with 3^+ valence and is a prime literature example of very good agreement between renormalized LDA band calculations and dHvA Fermi surface and effective mass measurements [4,5]. The focus of this work is the electronic structure and FS topology of $CeRu_2Si_2$ as measured by angle-resolved photoemission (ARPES). Previous characterization of the $4f^0$ $LaRu_2Si_2$ 4^+ valence system [6,7] and comparison to LDA calculations of both systems [8, 9] provide the framework for discussion and interpretation of the $CeRu_2Si_2$ results.

EXPERIMENT

The XRu_2Si_2 systems have the $ThCr_2Si_2$ crystal structure with a body-centered tetragonal Brillouin zone (BZ). A single crystal $CeRu_2Si_2$ sample was cleaved in ultra-high vacuum ($<6\times10^{-11}$ torr) at <130 K exposing the [001] surface. ARPES measurements were performed at ALS Beamline 7.0.1.2 in the photon energies range of 80-200 eV with a total instrumental resolution of ≈80 meV and full angular acceptance of ≈0.7°. The Fermi-edge intensity mapping technique using automated angular motions was employed to gain an overview of the sample orientation and Fermi surface structure. Valence spectra were then acquired along the [100] and [110] high symmetry azimuth directions for comparison to LDA band structure calculations. Additional ARPES experiments focused on the ***k***-dependent Ce 4*f* spectral weight, not discussed here, were performed at $4d\rightarrow4f$ resonant photon energies (122 eV) [6].

RESULTS

Fig. 1(a) shows reverse grayscale images of $CeRu_2Si_2$ valence band spectra at 154 eV, corresponding to the Γ-point at normal emission. A global comparison of this data to the LDA calculation in Fig. 1(b) shows very good qualitative agreement below 0.5 eV, especially with the parabolic bands centered on X and midway between Γ and Z with band minima at ≈1.6 eV binding energy. Also shown for comparison in Fig. 1(b) is the calculated band structure for $LaRu_2Si_2$ (dashed lines). The band theory comparison tells us that the introduction of relatively flat unoccu-

pied Ce 4*f* states ≈0.5-1 eV above E_F pushes bands 4 and 5 around and results in shallower dispersions and band-filling, *e.g.* smaller diameter hole-like E_F-crossings, consistent the extra *f*-electron in the Ce system relative to $LaRu_2Si_2$. Also experimentally observed in Fig. 1(a) is spectral weight at E_F over large regions of the FS which originates from Ce 4*f* states whose cross section is still relatively strong at 154 eV in spite of being 30 eV above the 4*d*-edge resonance at 122 eV. While this tells us something important about the 4*f* states, it obscures the details of the *d*-band FS crossings.

To further investigate the near-E_F *d*-band structure, we turn attention to the FS map in Fig. 2(a) acquired at 91 eV, which is below the 122 eV resonance so that the 4*f* states are weaker, and which is in a region of large *d*-cross section. This map was acquired over a 220° azimuth range out to 30° polar angle and has been 4-fold symmetrized. Also Fig. 2(b) shows calculated 2D FS contours for $CeRu_2Si_2$ with overplotted BZ dimensions and Γ, Z and X labels. Clearly observable features the experimental map include: *(i)* a bright intensity at the normal emission Γ-point, *(ii)* an intense closed contour in the second BZ's that correspond to near Γ-points, *(iii)* weak intensity and possibly small closed topologies around the Z-points. The existence of small closed FS pockets at the Γ and Z-points is in qualitative agreement with the theoretical contours.

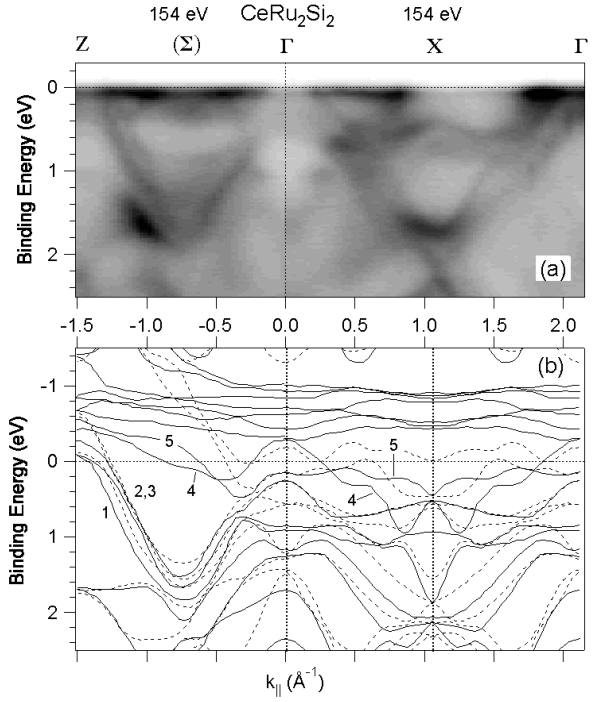
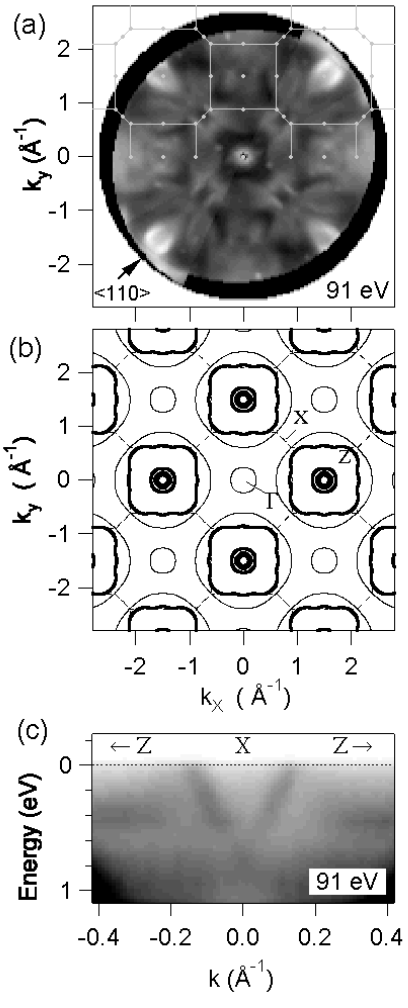


Figure 1. (a) Valence band intensity maps of $CeRu_2Si_2$ at 154 eV along high-symmetry azimuth angles corresponding to Γ−(Σ)−Z and Γ−X−Γ. (b) Theoretical band structure calculation for $CeRu_2Si_2$ (solid) and $LaRu_2Si_2$ (dashed).



(right) Figure 2. (a) Off-resonance Fermi-energy intensity maps of $CeRu_2Si_2$ at 91eV. (b) Theoretical contours of hole (bold) and electron (fine) Fermi surface topologies. (c) Valence band dispersion at the X-point at 91 eV orthogonal to Γ-X.

A weaker feature of great importance is the existence of a large circular contour centered on the second BZ Z-points and extending into the central BZ. The pairs of straight segments of intensity on either side of the X-points are parts of this large FS contour. At first glance, their similar sizes and shapes tempts one to associate the large Z-point contour in experiment with the large Z-point electron contour in theory. Apparently absent in this association is the "pillow" hole FS contour that closely follows the square BZ boundary centered on Z. Further consideration of the data suggests the reverse interpretation, that the theoretical electron surface is not observed and that the band 4 hole FS does appear in the data, but with a larger FS diameter more appropriate to $LaRu_2Si_2$ [see dashed lines in Fig. 1(b)] rather than to the smaller "pillow" contour calculated for $CeRu_2Si_2$. Strong support for this interpretation is provided by the great overall similarity of the experimental data for $LaRu_2Si_2$ [6,7] and $CeRu_2Si_2$, and by the spectra of Fig. 2(c) showing the dispersions that define the large surface near the X-point along the Z–X–Z direction. The spectra clearly show a single hole band (relative to Z) on each side of the X-point, as is appropriate for the single hole surface of the La compound, whereas one would observe two bands and two FS crossings for the adjacent hole and electron surfaces predicted for the Ce compound. Equivalent spectra for $LaRu_2Si_2$ are very similar in showing the single band.

CONCLUSION

The surprising finding is that, in contrast to dHvA results, we observe a Fermi surface for $CeRu_2Si_2$ that is essentially the same as that of the $4f^0$ La compound, thereby suggesting that the $4f^1$ electrons do not participate in the FS electron counting. This observation is consistent with a very deep conjecture put forth by Zwicknagl [4] that at temperatures far in excess of the Kondo temperature (T_K) where the magnetic moment is unquenched, the FS should exclude the *f*-electrons. We take note that the temperature of these measurements is six to seven times that of $T_K \approx 20K$ for $CeRu_2Si_2$, whereas the dHvA data are taken for low T (<3 K) in the Fermi liquid regime. The next challenge to confirm this interpretation is to compare the FS topologies of $CeRu_2Si_2$ at temperatures above and below T_K.

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